

### **REMARKS**

Reconsideration of this application is respectfully requested. Claims 1-50 and 55-73 are pending. Because claims 3, 6, 8, 44, 45, 47-50, 55-70, 72, and 73 have been withdrawn from consideration, only claims 1, 2, 4, 5, 7, 9-43, 46 and 71 are at issue.

### **Indefiniteness Rejection**

Claims 20, 24-26, and 71 have been rejected as indefinite due to the terms “cellulose III” and “Rx.”

Cellulose III is a term commonly used in the art. *See* Rydholm, Pulping Processes, p. 118 (a copy of which is attached). As discussed at page 2, lines 25-28, of the specification,

When cellulose is activated with liquid ammonia, crystalline cellulose III is formed. As well known in the art, crystalline cellulose III has a significantly different structure and properties than other crystalline cellulose structures, such as crystalline cellulose II. See, for example, Rydholm, *supra*; and Ott, Spurlin and Grafflin, *supra*.

Accordingly, one of ordinary skill in the art would readily recognize and understand what is meant by the term “cellulose III.”

At page 16, lines 12-19, the specification describes how to determine the Rx value of a cellulose pulp:

The Rx value for the sheet pulp was determined as follows. X-ray diffraction (Philips PW 1729 X-Ray Generator with a PW 1840 Diffractometer) was performed on the sheet pulp to determine peak intensities for native Cellulose I and mercerized Cellulose II fibers. The Rx value is the ratio of the peak diffraction for Cellulose II over the sum of the peak diffraction for Cellulose II and Cellulose I. See Rasch and McCarthy, *Norsk Skogindustri*, 8:329, 1954. The percent Cellulose II increases as the Rx value increases. The Rx value is derived from the intensities (I, counts per

second) of the diffracted radiation at approximately 12.6 and 15.6 degrees and is calculated according to the formula  $Rx = 2I_{12.6} / (2I_{12.6} + I_{15.6})$ .

Accordingly, contrary to the Examiner's assertion, the specification provides a definition for the term "Rx value."

For the foregoing reasons, the terms "cellulose III" and "Rx" are definite and applicants respectfully request withdrawal of this rejection.

### **Anticipation Rejection**

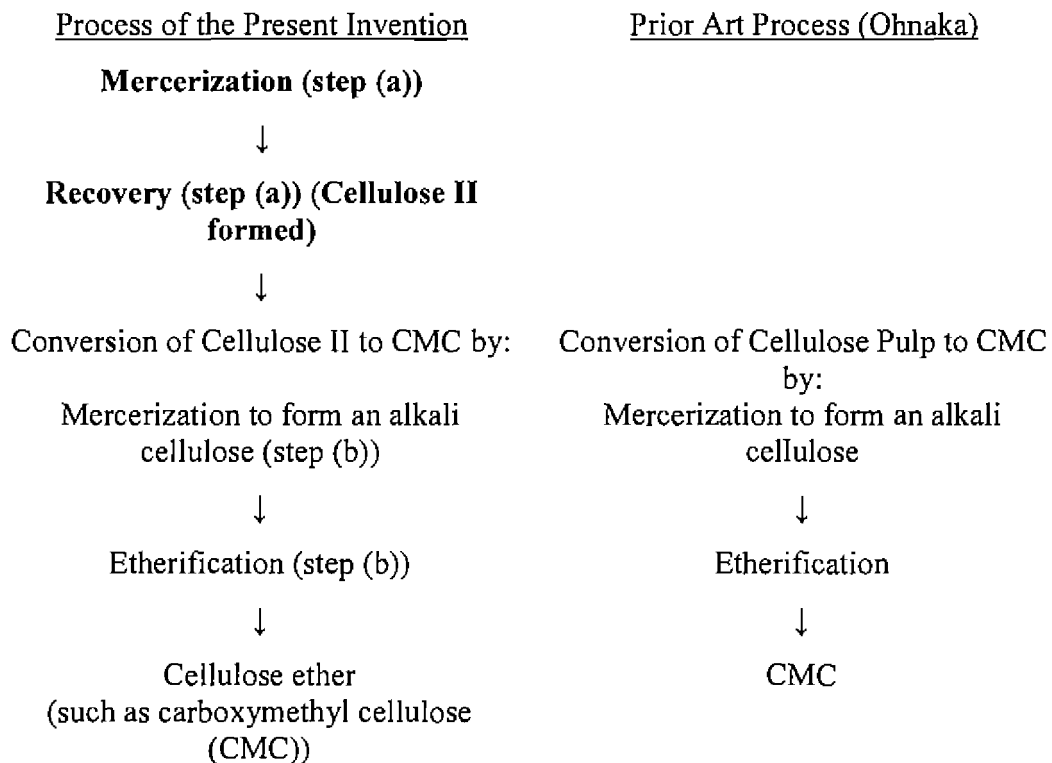
Claims 1, 10, 11, 14-19, 35-43, 46, and 71 have been rejected as anticipated by Ohnaka et al. (U.S. Patent No. 4,063,018). The Examiner contends that Comparative Examples 1-3 (col. 7-8) in Ohnaka anticipate these claims.

Applicants respectfully traverse this rejection and request reconsideration.

In the presently claimed process, the cellulose pulp is mercerized and recovered before being converted to a cellulose ether. Recovery involves removal or neutralization of most or all of the mercerizing agent (page 8, lines 3-4, of the specification). This converts the cellulose pulp from a first form, known as native cellulose or cellulose I, to a second crystal form, known as cellulose II. The cellulose II is then converted to a cellulose ether. For example, the cellulose ether can be formed by adding a mercerizing agent (e.g., sodium hydroxide) to form an alkali cellulose and then adding an etherifying agent (e.g., monochloroacetic acid). Thus, a mercerizing agent is used twice in the presently claimed process - once to pre-mercerize the cellulose pulp and once to form an alkali cellulose which is then etherified.

Below is a chart comparing the process of the present invention with that disclosed in Ohnaka. Ohnaka does not disclose or suggest mercerizing and recovering cellulose pulp before

converting it to a cellulose ether. In comparative examples 1-3 in Ohnaka, cellulose pulp was pulverized, sodium hydroxide was added to the pulverized cellulose pulp to form an alkali cellulose, and monochloroacetic acid was added to etherify the cellulose. A mercerizing agent (i.e., sodium hydroxide) was only added once, in contrast to the presently claimed process. Furthermore, in Ohnaka there is no washing or other recovery step following mercerization and prior to derivatization with monochloroacetic acid. As a result, the cellulose pulp which is used to form an alkali cellulose in Ohnaka is native cellulose (cellulose I), not cellulose II as in the presently claimed process.



For the foregoing reasons, Ohnaka does not anticipate the presently claimed process and applicants respectfully request withdrawal of this rejection.

**Obviousness Rejection**

Claims 2, 4, 5, 7, 9, 12, 13, and 20-34 have been rejected as obvious over Ohnaka in view of European Patent Publication No. EP 879,827 (EP '827).

Applicants respectfully traverse this rejection and request reconsideration.

A brief review of the chemistry and properties of cellulose and cellulose derivatives will put the invention of the claims in perspective and show how it is distinguishable from the prior art cited in the rejection. Mercerization is the swelling action of concentrated alkali, usually sodium hydroxide, on cellulose. Since cellulose is not soluble in water, the reaction is conducted as a heterogeneous reaction upon an aqueous slurry of cellulose fibers in water. The complete range of temperature and concentration conditions for mercerization with sodium hydroxide have been exhaustively studied by X-ray methods on mercerized and recovered cellulose. *See* Cellulose and Cellulose Derivatives, Volume V of HIGH POLYMERS, edited by Emil Ott, pages 272-345 on Mercerization, Interscience Publisher, Inc., New York, NY, 1943 (a copy of which is submitted herewith). *See especially*, Ott at 273-274.

Very interesting is a description of changes to the microscopic structure of cellulose fibers. In a section of Ott starting at page 338 written by E. I. Valko more than fifty years ago, the changes to cotton fiber were described as follows:

Immediately before the ripe boll burst open and exposes its flocks of cotton to the atmosphere, the fibers are tubular, with the hollow center (the "lumen") occupying a considerable portion of the cross section. No convolutions or twists are present, and the cross sections approach very closely to an elliptical or even circular form. On exposure to the air, the fiber dries out quickly and collapses to form a flat, convoluted ribbon. Commercial cotton hair is in this form.

When the cotton hair is brought into mercerizing solution, it begins to swell at once. In a few seconds it become elliptical, and, on further swelling, the cross section becomes almost circular. The diameter of this section is at least 25 to 30% greater than the width of the collapsed fiber.<sup>33</sup> . . . . During the mercerization process, the

cellulose of the walls swells inwardly. After the alkali treatment and during the washing and drying, the cross section shrinks. During this shrinkage, the hair retains the same circular form, but the shrinkage proceeds uniformly toward the axis and the lumen does not recover its original size. . . . By the strong swelling, the arrangement which was responsible for the collapse of the native hair is broken up, and the cylindrical form is retained upon subsequent drying.

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<sup>33</sup> M. A. Calvert and F. Summers, *J. Textile Inst.*, 16 T233 (1925).

Although many of the literature discussions of mercerization talk of it as being the treatment with alkali, it must be noted that physical studies of the effects of mercerization have all been performed on cellulose, which, after treatment with alkali, has been washed and dried. Washing removes the alkali. More recent discussions talk in terms of an irreversible change from native, amorphous cellulose I to cellulose II or crystalline cellulose.

Sarko et al. (a copy of which is submitted herewith) have studied the conversion of the crystal structure of native cellulose (cellulose I) to that of cellulose II through a series of crystalline intermediate alkali cellulose complexes. See Crystalline Alkali-Cellulose Complexes as Intermediates During Mercerization by Sarko et al, page 169-177 written as Chapter 9 of The Structures of Cellulose, Rajai H. Atalla, Editor, ACS Symposium Series Volume 340, American Chemical Society, Washington, DC, 1987. "Removing the NaOH from the structure through washing with water removes the energy-lowering electrostatic field. This results in a conversion of the structure to the only energy-lowering one that is available to it – a twofold helical, interchain hydrogen-bonded sheet structure." Sarko at page 176. While treatment with alkali to produce an alkali cellulose occurs in mercerization, it is the washing to remove the alkali which produces the final irreversible transformation to cellulose II.

"Mercerization is the name given to the conversion accomplished by swelling native cellulose fibres in concentrated sodium hydroxide solution. Although no dissolution occurs, the swelling allows for reorganization of the chains, and cellulose II results when the swelling agent is

removed.” Kolpak et al., *Polymer*, 19:123-131, 123 (Feb. 1978) (a copy of which is submitted herewith).

In the conventional derivatization process to form a carboxymethyl cellulose (CMC), the first step is the formation of an alkali cellulose, a heterogeneous reaction performed on a slurry of the starting cellulose with alkali (e.g., sodium chloride). This is followed by derivatization of the cellulose. Derivatization performed on an alkali cellulose produced from **native cellulose (cellulose I)** is described many times in the prior art and in the cited references used to reject the instant claims. However, that alkali cellulose does not contain cellulose II.

In contrast, the cellulose ether of this invention is produced by derivatization of **cellulose II**, which has been produced by mercerization and recovery of the pulp prior to formation of an alkali cellulose in the derivatization process. Recovery involves removal or neutralization of most or all of the mercerizing agent (page 8, lines 3-4, of the specification).

Neither Oknaka nor EP '827 disclose or suggest preparing cellulose ethers (such as CMC) by mercerizing and recovering cellulose pulp before converting it to the cellulose ether. Accordingly, a *prima facie* case of obviousness has not been established.

Additionally, the cellulose ethers, including CMC, produced by the presently claimed process exhibit significantly higher viscosities than those produced by the conventional etherification process. The viscosity of a CMC prepared by any process is dependent, *inter alia*, on the starting cellulose pulp. For example, CMC produced from cotton linters pulp generally has a higher viscosity than that prepared under the same process conditions from southern hardwood kraft pulp.

For any given starting cellulose pulp, the process of the present invention yields a CMC having a significantly higher solution viscosity than a CMC produced by the typical prior art method of preparing CMC by simply alkalating and etherifying cellulose pulp as described in Oknaka.

Tables 1-4 on the attached sheets show the viscosity of CMC's prepared as described in Examples 1-5, 7, 9, and 10 of the present application from various starting cellulose pulps, including never-dried and re-wetted cellulose pulps which had been converted from cellulose I to cellulose II by mercerization and recovery. The control for each example was prepared by the same procedure as the example on the native cellulose (cellulose I) which was not mercerized and recovered before being alkalated and etherified.

Tables 1-4 show that CMC's produced by the process of the present invention have significantly higher viscosities than CMC's produced by prior art processes. For example, Table 1 shows that when a cotton linter pulp starting material is mercerized and recovered prior to alkalating and etherifying, the viscosity of the CMC formed increases by at least 102%. Table 4 shows that when a sulfite pulp starting material is mercerized and recovered prior to alkalating and etherifying, the viscosity of the CMC formed increases by at least 104%.

For the foregoing reasons, Ohnaka alone or in combination with EP '827 does not render obvious the presently claimed invention and applicants respectfully request withdrawal of this rejection.

In view of the above remarks, applicants respectfully request that the application be reconsidered and that all pending claims be allowed and the case passed to issue. If there are any other issues remaining, the Examiner is respectfully requested to contact the undersigned attorney at the telephone number indicated below.

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Respectfully submitted,

By 

Jay P. Lessler

Registration No.: 41,151

DARBY & DARBY P.C.

P.O. Box 5257

New York, New York 10150-5257

(212) 527-7700

(212) 527-7701 (Fax)

Attorneys/Agents For Applicant



Table 1

CMC Derived from Cotton Linter Pulp				
Example No. in the present application	Concentration of NaOH (%)	1% Solution Viscosity (cP)	Control's 1% Solution Viscosity (cP)	Percentage Change in Viscosity
1 (Never-dried) <sup>1</sup>	14	80095	34690	130.89%
	18	83745	34690	141.41%
7 (Never-dried)	18 (Average)	79410	21493	269.47%
7 (Re-wetted dried)	18 (Average)	43453	21493	102.17%
9 (Never-dried)	18	75190	30145	149.43%
10 (Never-dried)	18	55335	25685	115.44%
10 (Never-dried)	18	57190	13920	310.85%

Table 2

CMC derived from Southern Softwood Kraft Pulp				
Example No. in the present application	Concentration of NaOH (%)	1% Solution Viscosity (cP)	Control's 1% Solution Viscosity (cP)	Percentage Change in Viscosity
2 <sup>2</sup>	10	607	586	3.58%
	14	1268	586	116.38%
	18	1147	586	95.73%
3 (Never-dried)	7	679	550	23.45%
	10	1108	550	101.45%
	14	1365	550	148.18%
	18	1472	550	167.64%

<sup>1</sup> The 7 and 10% NaOH treated pulp contained 0% sheet cellulose II and, therefore, was not mercerized.

<sup>2</sup> Table 2 in the specification indicates that the 7% NaOH treated pulp contained 0% sheet cellulose II and, therefore, was not mercerized..

Table 3

CMC derived from Southern Hardwood Kraft Pulp				
Example No. in the present application	Concentration of NaOH (%)	1% Solution Viscosity (cP)	Control's 1% Solution Viscosity (cP)	Percentage Change in Viscosity
5 <sup>3</sup>	10	461	230 or 362	100.43% or 27.35%
	14	547	230 or 362	137.83% or 51.10%
	18	923	230 or 362	301.30% or 154.97%

Table 4

CMC derived from Northern Softwood Sulfite Pulp				
Example No. in the present application	Concentration of NaOH (%)	1% Solution Viscosity (cP)	Control's 1% Solution Viscosity (cP)	Percentage Change in Viscosity
4 <sup>4</sup>	10	2037	898 or 1087	126.84% or 87.40%
	14	1835	898 or 1087	104.34% or 68.81%
	18	2608	898 or 1087	190.42% or 139.93%

<sup>3</sup> Table 6 in the specification indicates that the 7% treated pulp contained only 1% sheet cellulose II.

<sup>4</sup> Table 5 in the specification indicates that the 7% treated pulp contained 0% sheet cellulose II and, therefore, was not mercerized.